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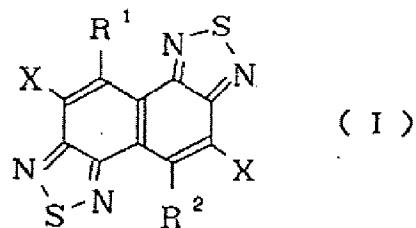
[Document Name]Description

[Title of the Invention]An organic electroluminescence element

[Claim(s)]

[Claim 1]In an organic electroluminescence element by which an anode, an organic electron hole transportation layer, an organic electron transport layer, and the negative pole were laminated one by one, An organic electroluminescence element, wherein an organic electron hole transportation layer and/or an organic electron transport layer contain naphth ****- 1 and 2 denoted by following general formula (I), and 5-thiadiazole derivative.

[Chemical formula 1]



the inside of a formula, R¹, and R² -- a hydrogen atom, a halogen atom, and a cyano group. A nitro group, a carboxyl group, an alkyl group that may have a substituent, An aralkyl group which may have a substituent, an alkenyl group which may have a substituent, An amino group which may have a substituent, an amide machine which may have a substituent, An alkoxy group which may have a substituent, an alkoxy carbonyl group which may have a

substituent, A heterocyclic machine which may have an aromatic hydrocarbon machine or a substituent which may have an alkoxy sulfonyl group which may have a substituent, and a substituent is shown, and X shows a hydrogen atom, a halogen atom, an alkoxy group, or a hydroxyl machine.

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to an organic electroluminescence element. It is related with the thin film type device which emits light applying ** and an electric field to the combination of the electron hole transportation layer and electron transport layer which comprise an organic compound in detail.

[0002]

[Description of the Prior Art] Although what doped Mn which is a luminescence center, and rare earth elements (Eu, Ce, Tb, Sm, etc.) is common to ZnS and CaS which are the II-VI fellows compound semiconductors of the charge of non-equipments, SrS, etc. as a thin film type electroluminescence element conventionally, As for the electroluminescence element produced from the above-mentioned charge of non-equipments, one alternating current drive has the problem that necessity (50-1000 Hz) and 2 drive voltage are high (-200V) and that the formation of 3 full color has the high cost of difficulty (especially blue is a problem) and 4 circumference drive circuit.

[0003] However, development of the electroluminescence element using organic materials came to be performed in recent years for improvement of the above-mentioned problem. [besides the anthracene known from before as a luminous layer material, pyrene, etc.] a cyanine pigment (J. Chem. Soc. and Chem. Commun. -- 557 pages) 1985 and PIRAZORIN (Mol. Crys. Liq. Cryst. -- 135 volumes) PERIREN (Jpn. J. Appl. Phys., 25 L 773 pages, 1986)

or a coumarin series compound, tetra-phenyl butadiene (JP,S57-51781,A), etc. are reported in 355 pages and 1986.

[0004]It aims at improvement in the pouring efficiency of Carrier from an electrode in order to raise luminous efficiency, Optimization of an electrode kind, the device (JP,S57-51781,A, JP,S59-194393,A, JP,S63-295695,A, Appl. Phys. Lett., 51 volumes, 913 pages, 1987) which provides the luminous layer which consists of an electron hole transportation layer and an organic fluorescent substance, etc. are performed. [the purpose of changing a luminescence color while raising the luminous efficiency of an element] Doping fluorescence pigments for laser, such as Kumarin, by making the aluminium complex of 8-hydroxy quinoline into host material (65 volumes of J. Appl. Phys., 3610 pages, 1989) is also performed.

[0005]

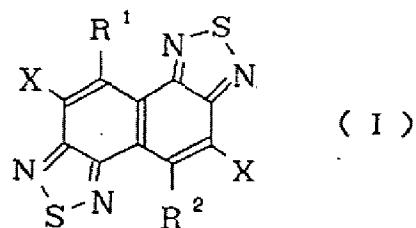
[Problem to be solved by the invention]However, the organic electroluminescence element of luminescence performance, especially luminous efficiency currently indicated by these is still insufficient, and the further improvement examination was desired. As a result of inquiring wholeheartedly for the purpose of providing the organic electroluminescence element which can be made to drive stably over a long period of time in view of the above-mentioned actual condition, this invention persons found out that a specific compound was preferred, and completed this invention.

[0006]

[Means for solving problem]Namely, in the organic electroluminescence element by which, as for the gist of this invention, an anode, an organic electron hole transportation layer, an organic electron transport layer, and the negative pole were laminated one by one, An organic electron hole transportation layer and/or an organic electron transport layer consist in naphth ****- 1 and 2 denoted by following general formula (I), and the organic electroluminescence element containing 5-thiadiazole derivative.

[0007]

[Chemical formula 2]



[0008]the inside of a formula, R^1 , and R^2 -- a hydrogen atom, a halogen atom, and a cyano group. A nitro group, a carboxyl group, the alkyl group that may have a substituent, The aralkyl group which may have a substituent, the alkenyl group which may have a substituent, The amino group which may have a substituent, the amide machine which may have a substituent, The alkoxy group which may have a substituent, the alkoxy carbonyl group which may have a substituent, The heterocyclic machine which may have the aromatic hydrocarbon machine or substituent which may have the alkoxy sulfonyl group which may have a substituent, and a substituent is shown, X shows a hydrogen atom, a halogen atom, an alkoxy group, or a hydroxyl machine. The organic electroluminescence element of this invention is hereafter explained according to an accompanying drawing.

[0009]Drawing 1 is a sectional view showing typically the constructional example of the organic electroluminescence element of this invention, as for 1, a substrate, and 2a and 2b express an electric conduction layer, 3 expresses an organic electron hole transportation layer, and 4 expresses an organic electron transport layer respectively. Although the substrate 1 serves as a base material of the organic electroluminescence element of this invention and the board of quartz or glass, a metal plate, metallic foil and a plastic film, a sheet, etc. can be used, Transparent synthetic resin boards, such as a glass board, polyester, poly methacrylate, polycarbonate, poly SARUHON, are preferred.

[0010]Although the electric conduction layer 2a is formed on the substrate 1, This electric conduction layer 2a is usually constituted by conductive resin, such as metal oxides, such as

an oxide of metal, such as aluminum, gold, silver, nickel, palladium, and TERURU, indium, and/or tin, copper iodide, carbon black, or poly (3-methylthiophene), etc. Although formation of the electric conduction layer 2a is usually performed by the sputtering method, vacuum evaporation method, etc. in many cases, In the case of metal particulates, such as silver, or copper iodide, carbon black, conductive metal oxide particulates, conductive resin fine powder, etc., it can also form by distributing in suitable binder resin solution and applying on a substrate. In the case of conductive resin, a thin film can also be directly formed on a substrate by electric field polymerization. The electric conduction layer 2a can also laminate and form a different substance.

[0011]Although the thickness of the electric conduction layer 2a changes with transparency to need, when transparency is needed, it is desirable for the transmissivity of visible light to be not less than 80% preferably not less than 60%, and 50-10000 A of thickness is usually about 100-5000A preferably in this case. When it may be opaque, the electric conduction layer 2a may be the same as that of the substrate 1. The electric conduction layer 2a can also be laminated by a different substance.

[0012]In the example of drawing 1, the electric conduction layer 2a plays the role of electron hole pouring as an anode (anode). On the other hand, the electric conduction layer 2b plays the role which injects an electron into the organic electron transport layer 4 as the negative pole (cathode). Although the material used as the electric conduction layer 2b can use the material for said electric conduction layers 2a, in order to perform electronic pouring efficiently, metal with a low value of a work function is preferred, and metal or those alloys, such as tin, magnesium, indium, aluminum, and silver, are used. The film thickness of the electric conduction layer 2b is comparable as the electric conduction layer 2a, and can usually be formed by the same method as the electric conduction layer 2a.

[0013]Although not shown in drawing 1, the still more nearly same substrate as the substrate 1 can also be provided on the electric conduction layer 2b. However, as an electroluminescence element, at least one side of the electric conduction layer 2a and the electric conduction layer 2b needs the good thing of transparency. From this, as for one side of the electric conduction layer 2a and the electric conduction layer 2b, it is preferred that it is 100-5000-A film thickness, and a good thing of transparency is desired.

[0014]Although the organic electron hole transportation layer 3 is formed on the electric conduction layer 2a, the organic electron hole transportation layer 3 is formed from the compound which can convey the electron hole from an anode in the direction of the organic electron transport layer 4 efficiently in inter-electrode [which was able to give the electric field]. As an organic electron hole transportation compound, the electron hole pouring efficiency from the electric conduction layer 2a is high, and it is required to be a compound which can convey the poured-in electron hole efficiently. For that purpose, ionization potential is small, and moreover hole mobility is large, and also it is required that the impurities used as the trap excellent in a stabilization positive should be compounds which are hard to generate at the time of manufacture and use.

[0015]As such an electron hole transportation compound, what is written in the 13-14th columns of the 5-6th page and U.S. Pat. No. 4175960 of JP,S59-194393,A is mentioned, for example. As a desirable example of these compounds, the N and N'-diphenyl N, N'-(3-methylphenyl)-1, and 1'-biphenyl 4, 4'-Gia Min, Aromatic amine system compounds, such as a 1 and 1'-bis(4-G p-tolylamino phenyl)cyclohexane, 4, and 4'-bis(diphenylamino)KUWADORO phenyl, are mentioned. Except an aromatic amine system compound, the hydrazone compound shown in JP,H2-311591,A is mentioned. These aromatic amine compounds or a hydrazone compound may be respectively used as a mixture if needed, even if it uses independently.

[0016]The organic electron hole transportation layer 3 is formed by laminating on said electric conduction layer 2a by the applying method or a vacuum evaporation method. For example, the binder resin from which it does not become a trap of an electron hole according to one sort or two sorts or more, and necessity about an organic electron hole transportation compound in the case of the applying method, The application solution which added additive agents, such as application nature improvement agents, such as a REPERINGU agent, and in which it was made to dissolve is adjusted, and it applies on the electric conduction layer 2a by methods, such as a spin coat method, and it dries and the organic nature hole transportation layer 3 is formed. Polycarbonate, poly acrylate, polyester, etc. are mentioned as binder resin. Since it will reduce hole mobility if binder resin has many amounts of addition, few directions are desirable and its 50 or less weight % is preferred.

[0017]After paying organic electron hole transportation ingredients to the crucible installed in the vacuum vessel and exhausting the inside of a vacuum vessel even to 10^{-6} Torr with a suitable vacuum pump, in the case of a vacuum evaporation method, a crucible is heated, electron hole transportation material is evaporated in it, and a layer is formed on the substrate which faced the crucible and was placed. 100-3000 A of film thickness of the organic electron hole transportation layer 3 is usually 300-1000A preferably. Thus, in order to form a thin film uniformly, a vacuum evaporation method is used well.

[0018]Although the organic electron transport layer 4 is formed on the organic electron hole transportation layer 3, the organic electron transport layer 4 is formed from the compound which can convey the electron from the negative pole in the direction of the organic electron hole transportation layer 3 efficiently in inter-electrode [which was able to give the electric field]. As an organic electron transport compound, the electronic pouring efficiency from the electric conduction layer 2b is high, and it is required to be a compound which can convey the poured-in electron efficiently. For that purpose, it is required that the impurities which electron affinity is large, and the degree of electron transfer is moreover large, and also are excellent in stability, and serve as a trap should be compounds which are hard to generate at the time of manufacture and use.

[0019]As a material which fulfills such conditions, aromatic compounds, such as tetra-phenyl butadiene (JP,S57-51781,A), Metal complexes, such as an aluminium complex of 8-hydroxy quinoline (JP,S59-194393,A), A cyclo PENTA diene derivative (JP,H2-289675,A), a PERINON derivative (JP,H2-289676,A), an oxadiazole derivative (JP,H2-216791,A) and a screw styryl benzene induction object (JP,H1-245087,A.) a 2-222484 gazette and a PERIREN derivative (JP,H2-189890,A.) a 3-791 gazette and a coumarin compound (JP,H2-191694,A.) A 3-792 gazette, a rare earth complex (JP,H1-256584,A), a JISUCHIRIRU pyrazine derivative (JP,H2-252793,A), a thiadiazolo pyridine derivative (JP,H3-37292,A), a pyrrolo pyridine derivative (JP,H3-37293,A)

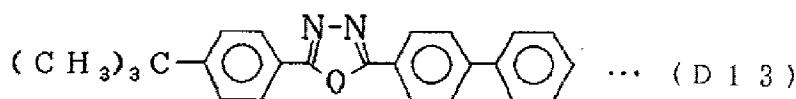
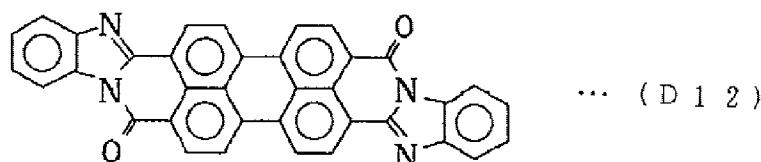
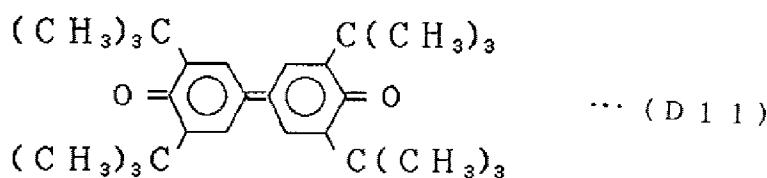
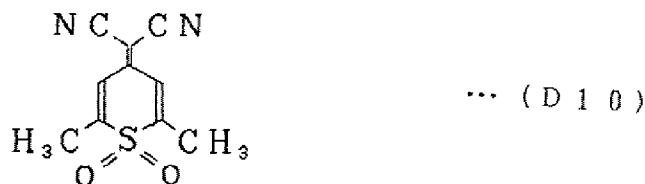
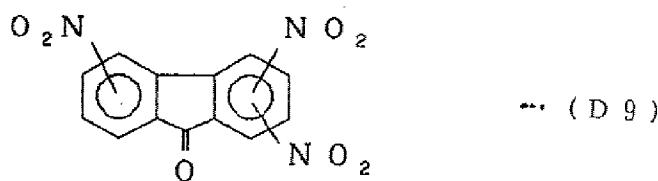
A NAFUCHI lysine derivative (JP,H3-203982,A) etc. are mentioned. When these compounds are used, the organic electron transport layer 4 plays simultaneously the role which conveys an electron, and the role which brings about luminescence in the case of re-combination of an electron hole and an electron.

[0020]100-2000 A of film thickness of the organic electron transport layer 4 is usually 300-

1000A preferably. Although the organic electron transport layer 4 can also be formed by the same method as the organic electron hole transportation layer 3, a vacuum evaporation method is usually used. In order to raise the luminous efficiency of an organic electroluminescence element more, as shown in drawing 2, other organic electron transport layers 5 can also be laminated on the organic electron transport layer 4. It is required that electronic pouring from the negative pole should be easy for the compound used for this organic electron transport layer 5, and electronic transport capacity should be still larger. Nitroglycerine substitution fluorenone derivatives, such as a compound denoted by the following structural formula (D9) as such an organic electron transport compound, Thiopyran dioxide derivatives, such as a compound denoted by the following structural formula (D10), Diphenyl quinone derivatives, such as a compound denoted by the following structural formula (D11), PERIREN tetracarboxylic acid derivatives (Jpn. J. Appl. Phys. -- 27 volumes), such as a compound denoted by the following structural formula (D12) Oxadiazole derivatives (Appl.Phys.Lett.55 volume, 1489 pages, 1989), such as a compound denoted by the following structural formula (D13), will be mentioned in L269 page and 1988.

[0021]

[Chemical formula 3]



[0022] 100-2000 Å of such film thickness of the organic electron transport layer 5 is usually 300-1000 Å preferably. It is also possible to laminate on a structure contrary to drawing 1, i.e., a substrate, in order of the electric conduction layer 2b, the organic electron transport layer 4, the organic electron hole transportation layer 3, and the electric conduction layer 2a, and at least one side is able to provide the electroluminescence element of this invention between two substrates with high transparency, as mentioned already. It is possible similarly to use structure contrary to drawing 2.

[0023]In order to change a luminescence color, while raising the luminous efficiency of an organic light emitting element conventionally, doping various kinds of fluorescence pigments

by making the aluminium complex of 8-hydroxy quinoline into host material is performed -- **** (U.S. Pat. No. 4,769,292 item). as the advantage of the method by this dope -- (1) -- the fluorescence pigment which luminous efficiency investigates with an efficient fluorescence pigment, and a luminescence wavelength investigates by improvement and selection of (2) fluorescence pigment and which starts (3) concentration quenching -- usable and (4) -- usable and ** are mentioned also for a bad filmy fluorescence pigment.

[0024]In this invention, when an organic electron transport layer plays the role, as a host material, the above-mentioned organic electron transport compound is mentioned, and when an organic electron hole transportation layer plays the role, as a host material, an above-mentioned aromatic amine compound and hydrazone compound are mentioned. In this invention, the fields where the compound denoted by said general formula (I) is doped may be the organic electron hole transportation layer 3 and/or the organic whole electron transport layer 4, or may be the part. 10^{-3} - 10-mol% of the quantity by which the compound denoted by said general formula (I) is doped to host material is desirable.

[0025]When strong fluorescence is shown in the state of solution and it is doped by host material, the luminous efficiency of the compound denoted by said general formula (I) of an element improves. Since the filmy voice of host material can be stabilized structurally, it is possible to give the stability over a long period of time to an organic electroluminescence element. [the compound denoted by said general formula (I)] [as a method of doping to the organic electron hole transportation layer 3 and/or the organic electron transport layer 4] In the case of the applying method, with for example, an organic electron hole transportation compound or an organic electron transport compound, the compound denoted by said general formula (I), and also necessity, The application solution which added additive agents, such as binder resin which does not become a trap of an electron hole or an electron as occasion demands, and application nature improvement agents, such as a REPERINGU agent, and in which it was made to dissolve is adjusted, and it applies by methods, such as a spin coat method, and dries and forms. Polycarbonate, poly arylate, polyester, etc. are mentioned as binder resin. Since it will reduce the mobility of an electron hole or an electron if binder resin has many amounts of addition, few directions are desirable, and its 50 or less weight % is preferred. In the case of a vacuum evaporation method, an organic electron hole transportation compound or an organic electron transport compound is put into the crucible installed in the vacuum vessel, After putting the compound denoted by said general formula (I) into another crucible and exhausting the inside of a vacuum vessel even to a 10^{-6} -Torr grade with a suitable

vacuum pump, each crucible is heated simultaneously, and is evaporated and a layer is formed on the substrate which faced the crucible and was placed. As another method, what mixed the above-mentioned material at a predetermined rate may be evaporated using the same crucible.

[0026]In said general formula (I), [as R¹ and R²] Preferably A hydrogen atom, a chlorine atom, a bromine atom, iodine Hitoshi Harako's halogen atom, Substitution or unsubstituted alkenyl groups, such as a cyano group, a nitro group, a carboxyl group or an allyl group, and 2-chloro allyl group, amide machine;CONH₂, CONHR, and CONRR' (the inside of a formula, and

R.) R' shows an aromatic hydrocarbon machine or the alkyl groups which may be replaced, such as a phenyl group. The amino group which may have a substituent, the methyl group which may have a substituent, The alkyl group of the carbon numbers 1-6, such as an ethyl group; The methoxy group which may have a substituent, The alkoxy group of the carbon numbers 1-6, such as an ethoxy basis; The methoxy carbonyl group which may have a substituent, The alkoxy carbonyl group of the carbon numbers 1-6 of an ethoxycarbonyl machine etc.; The methoxy sulfonyl group which may have a substituent, Alkoxy sulfonyl groups, such as an ethoxy sulfonyl group; The benzyl group which may have a substituent, Aralkyl groups, such as a FENECHIRU machine; The thienyl group which may have aromatic hydrocarbon machine; substituents which may have a substituent, such as a phenyl group, a naphthyl group, an acenaphthyl machine, and an anthryl group, a carbazol group, the India Lil machine, Heterocyclic machines, such as a frill machine, etc. are shown. As a substituent replaced by these, lower alkoxy group; phenoxy groups, such as an alkyl-group; methoxy group of the carbon numbers 1-6, such as a methyl group and an ethyl group, aryloxy group [, such as a bird OKISHI machine,]; -- ARIRU alkoxy group [, such as a benzyloxy group,]; -- substitution amino groups, such as aryl group; dimethylamino groups, such as a phenyl group and a naphthyl group, etc. are mentioned. A hydrogen atom, chlorine Hitoshi Harako's halogen atom, the alkyl group of the carbon numbers 1-6, and the alkoxy group of the carbon numbers 1-6 are chosen especially preferably.

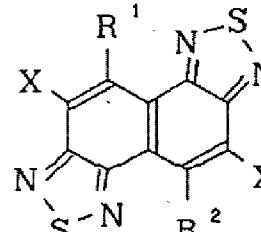
[0027]As X, the alkoxy group of the carbon numbers 1-6, such as a hydrogen atom, a chlorine atom, a bromine atom, iodine Hitoshi Harako's halogen atom, a methoxy group, and an ethoxy basis, a hydroxyl machine, etc. are mentioned preferably. the synthetic method of these compounds -- 18 volumes of J[Mataga and others]. Heterocyclic. Chem., and 1073page (1981); -- it is shown in said 26 volume and 215 pages (1989). Thus, the example of the above-mentioned compound obtained is illustrated below. These compounds have strong

fluorescence intensity and lightfastness and their heat resistance are also good.

[0028]

[Table 1]

表 1

		
化合物 N o.	R ¹ 、R ²	X
1	- H	- H
2	- H	- B r
3	- H	- C l
4	- H	- I
5	- H	- O H
6	- H	- O C H ₃
7	- B r	- B r
8	- C l	- C l

[0029]

[Function] In this invention, the fluorogenic compound denoted by said general formula (I) is

used as a dope material of an organic electron transport layer and/or an organic electron hole transportation layer.

Therefore, it becomes possible to bring about the outstanding luminescent property and the stability over a long period of time.

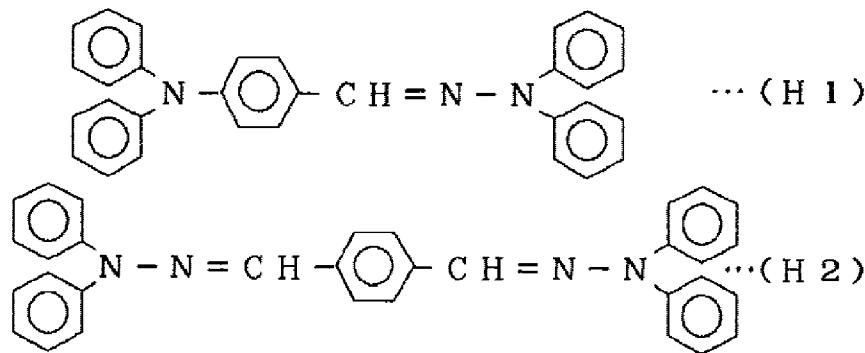
[0030]

[Working example] Next, although a work example explains this invention still more concretely, this invention is not limited to the description of the following work examples, unless the gist is exceeded. The organic electroluminescence element of the structure shown in work-example 1 drawing 1 was produced by the following methods.

[0031] After cleaning ultrasonically with organic alkali what deposited 1200A of indium-tin-oxide (ITO) transparent conductive films on the glass substrate with ultrasonic cleaning, a flush, and isopropyl alcohol, it installs in a vacuum evaporation system, It exhausted using the oil diffusion pump until the degree of vacuum in a device became below 2×10^{-6} Torr. What mixed the hydrazone compound denoted by the following structural formula (H1) and (H2) by the molar ratio as an organic electron hole transportation layer material 1:0.3,

[0032]

[Chemical formula 4]

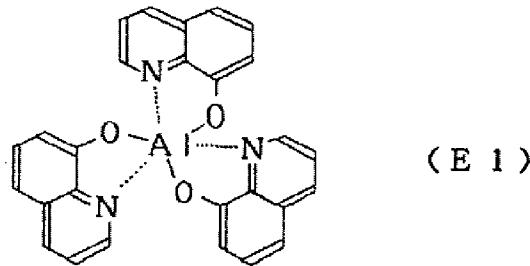


[0033]As a compound to dope, respectively, above naphth ****- 1 and 2 and 5-thiadiazole derivative (compound No.2) were put into another ceramic crucible, were simultaneously heated with Ta line heater around a crucible, and were evaporated in the vacuum vessel. To the hydrazone mixture, the range of the temperature of a crucible is 130-150 **, and it was controlled in 90-110 ** to naphth ****- 1 and 2 and 5-thiadiazole derivative. The degree of vacuum at the time of evaporation was 1×10^{-6} Torr. thus -- compound No.2 receives the above-mentioned hydrazone mixture -- 3-mol % -- the doped organic electron hole pouring transportation layer was vapor-deposited by 514-A film thickness. Vapor deposition time was 3 minutes.

[0034]Next, the 8-hydroxy quinoline complex of the aluminum denoted by the following structural formula (E1) is used as a material of an organic electron transport layer,

[0035]

[Chemical formula 5]



[0036]It vapor-deposited by controlling the temperature of a crucible in 205-230 ** like the organic electron hole transportation layer. The degree of vacuum at the time of vapor deposition was 7×10^{-7} Torr, and vapor deposition time was 2 minutes. As a result, the organic electron transport layer of 520 A of film thickness was obtained. Finally the alloy electrode of 2 yuan of magnesium and silver was vapor-deposited to 1500 A of film thickness by the simultaneous vapor-depositing method as a cathode. The degree of vacuum of vapor

deposition was 6×10^{-6} Torr using the molybdenum boat, and vapor deposition time was 8 minutes. The glossy film was obtained. The range of the atomic ratio of magnesium and silver was 10:3.

[0037]Thus, the result of the luminescent property which impressed and measured the direct-current voltage of minus to plus, and magnesium and a silver electrode (cathode) at the ITO electrode (anode) of the produced organic electroluminescence element is shown in Table 2. This element showed uniform green luminescence. The organic electroluminescence element was produced like the work example 1 except not having doped compound No.2 in a comparative example 1 organicity electron hole transportation layer. The measurement result of the luminescent property of this element is shown in Table 2. This element showed uniform green luminescence.

[0038]

[Table 2]

表 2

素子	V _{th} [V]	最大輝度 [cd/m ²]	発光効率 [lm/W]
実施例 1	7	2 7 6 0	0. 3 1
比較例 1	1 0	1 4 7 1	0. 1 7

V_{th} : 輝度が 1 cd/m² となる電圧

[0039]Each element produced by the work example 2, the comparative example 2 work example 1, and the comparative example 1 is saved in a vacuum, and the result of having

measured the variation per hour of the practical use drive voltage (V_{100}) from which luminosity becomes 100 cd/m^2 is shown in drawing 3. When the element of the comparative example 1 passed the 30th day or more to the element of the work example 1 having shown the stability over a long period of time, while the increase in drive voltage became remarkable, luminosity also fell greatly.

[0040]

[Effect of the Invention]As for the organic electroluminescence element of this invention, an anode (anode), an organic electron hole pouring transportation layer, an organic electronic pouring transportation layer, and the negative pole (cathode) are provided one by one on the substrate.

Since the specific fluorescence pigment is doped to an organic electron hole pouring transportation layer and/or an organic electronic pouring transportation layer, or its part, When voltage is impressed by using both electric conduction layers as an electrode, even after being able to obtain luminescence of luminosity practically sufficient on low drive voltage and saving for a long period of time, early luminescent property can be maintained.

[0041]Therefore, the light source in which the electroluminescence element of this invention employed the feature as the field and field photogen of a flat panel display (for example, flat TV) efficiently. The application to (for example, the light source of a copying machine and the backlight light source of a liquid crystal display or instruments), a display board, and a beacon light can be considered, and the technical value is large.

[Brief Description of the Drawings]

[Drawing 1]It is a sectional view of one work example of the organic electroluminescence element of this invention.

[Drawing 2]It is a sectional view of the work example of others of the organic

electroluminescence element of this invention.

[Drawing 3]It is a figure showing the result of having measured the variation per hour of the practical use drive voltage of the organic electroluminescence element produced by the work example and comparative example of this invention.

[Explanations of letters or numerals]1 Substrates [Organic electron transport layer which comprises a compound which is different in 4] 2a and 2b Electric conduction layer 3 Organic electron hole transportation layer 4 Organic electron transport layer 5

[Translation done.]